Towards Optimized Electrolyte Formulations for Radical Polymer-based Dual-Ion Batteries

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Lithium ion batteries are unrivaled in terms of specific energy or energy density. However, the fulfillment of other properties like enhanced sustainability, safety, abundancy of the used materials and lower costs have to be brought into focus nowadays due to crucial importance of environmental and climate issues. A promising approach to head towards a more environmental friendlier battery is the so-called dual-ion battery (DIB), in which anions, besides cations, participate in the storage mechanism of the battery.[1],[2] This characteristic of the DIB allows the use of transition metal-free and therefore less toxic active materials like graphite or polymers.[3],[4]

Redox active polymer electrodes possess high opportunities for various applications, due to the easy manufacturing and tailoring of their redox properties. Furthermore, cells containing polymer electrodes show high rate capabilities because of their simple working principle in contrast to complex interaction mechanism in inorganic electrode materials.[4] Especially, polymers with isolated redox-active units are promising candidates, because of the elimination of the challenges resulting from conjugated polymer chains, e.g., voltage sloping during ongoing operation. In this study poly(2,2,6,6-tetramethylpiperidinyl-N-oxyl methacrylate) (PTMA) was used because of its favourable electrochemical properties and the high stability of its radical functional group.[4]

Therefore, this work aims at the optimization of the performance of PTMA positive electrodes in a DIB setup by using different electrolyte compositions. Cyclic voltammetry measurements show the redox behaviour and PTMA dissolution problematic in dependency of the used electrolyte solvents. Thereby, electrolytes with higher amounts of cyclic carbonates show enhanced cycling performance. Further, γ-butyro lactone (GBL) has been chosen as electrolyte solvent for systematic investigations, because of the possibility of the use as a single solvent in contrast to ethylene carbonate, which is solid at room temperature. In addition, GBL-based electrolytes show improved performance and higher rate capabilities at increased C-rates (above 2C) compared to 1M $LiPF_6$ in EC:EMC (3:7 by wt.).

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References:

[1] R. Schmuch, R. Wagner, G. Hörpel, T. Placke, M. Winter *Nat. Energy* 2018, **3**, 267–278.
[2] T. Placke, R. Kloepsch, S. Dühnen and M. Winter, *J. Solid State Electrochem.*, 2017, **21**, 1939-1964.
[3] T. Placke, A. Heckmann, R. Schmuch, P. Meister, K. Beltrop, M. Winter *Joule* 2018, **2**, 2528–2550.

[4] S. Muench, A. Wild, C. Friebe, B. Häupler, T. Janoschka, U. S. Schubert *Chem. Rev.* 2016, **116**, 9438-9484.